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(54) **Silver halide photographic material**

(57) A silver halide photographic light-sensitive material is disclosed comprising on a support;
 (1) silver halide grains containing at least 50 mole% of silver chloride;

(2) a liquid paraffin or finely particulate solid paraffin having an average particle size of not more than 5μ ; and
 (3) a compound comprising an alkylene oxide chain present in one or more layers.

GB 2 122 764 A

SPECIFICATION

Silver halide photographic light-sensitive material

BACKGROUND TO THE INVENTION

The present invention relates to a silver halide photographic light-sensitive material, and more particularly to a silver halide photographic light-sensitive material capable of producing a satisfactory photographic characteristics, especially an excellent contrast when processed in a lith-type developer liquid as well as in a non-lith-type developer liquid.

Silver halide photographic light-sensitive materials are generally used over a very wide field because of the ease of adjusting the photographic speed, gradation, etc., thereof.

First of all, the necessity in the lithographic field requires the application of a light-sensitive material having a considerably high contrast, so that there has been generally practiced such a method that, as an inherent technique, a silver halide photographic light-sensitive material containing a relatively large amount of silver chloride is used which is processed in a relatively low sulfite concentration-having developer comprised principally of a dihydroxybenzene-type compound (hereinafter called a lith-type developer) to thereby form an image consisting of halftone dots and lines of extremely high contrast.

Further, if the silver halide emulsion layer of the above-described silver halide photographic light-sensitive material contains a compound having in the molecule thereof a polyalkylene oxide chain as described in, e.g., Japanese Patent Examined Publication Nos. 25201/1967 and Japanese Patent Publication Open to Public Inspection (hereinafter referred to as Japanese Patent O.P.I. Publication) No. 108130/1977, then the image becomes further extremely contrasty so that the image quality is much improved.

As the light-sensitive material for graphic arts use wherein a high contrast of said image is required, satisfactory physical properties such as dimensional stability, adherence of the layer to the support, adaptability to the processing in an automatic processor, and the like, are required. In order to meet such requirements, the light-sensitive material is designed so that the amount of gelatin used therein is as small as possible.

However, the image quality improving effect by the abovementioned polyalkylene oxide-type compound depends largely upon the amount of the gelatin used. If the amount of gelatin is small, the effect becomes also fairly small, so that, in order to make up for the effect, the adding amount of the compound in a considerable amount is needed, the amount being in some cases required to be even as much as several grams per mole of silver halide.

As a processing liquid for silver halide light-sensitive material with such high contrast, on the other hand, there has also been used frequently a non-lith-type developer liquid which, although hardly capable of producing as much a contrasty image as the lith-type developer does, contains a relatively large amount of a sulfite for the stability thereof and a developing agent not restricted to a dihydroxybenzene-type compound (this combination is more generally used; the lith-type developer liquid is rather considered to be special), but if the light-sensitive material containing a large amount of the abovementioned alkylene oxide-type compound is processed in a developer of this type, the image contrast of the light-sensitive material becomes remarkably reduced with increasing fog, so that no good photographic characteristics can be obtained.

SUMMARY OF THE INVENTION

Accordingly, it is an object of the present invention to provide a silver halide photographic light-sensitive material which is capable of displaying satisfactory contrast when processed in either the lith-type or the non-lith-type developer liquid.

It is another object of the present invention to provide a silver halide photographic light-sensitive material which is capable of displaying an excellently contrasty image with the photographic speed and fog thereof not affected.

Other objects of the present invention will become apparent from the following descriptions of the invention and in the examples thereof.

According to the present invention, the above-described objects of the invention can be accomplished by a silver halide photographic light-sensitive material comprising on the support thereof at least one layer having therein (1) a silver halide grains containing at least 50 mole% of silver chloride; (2) a liquid paraffin or finely particulate solid paraffin having an average particle size of not more than 5 μ ; and (3) a compound having in the molecule thereof an alkylene oxide chain.

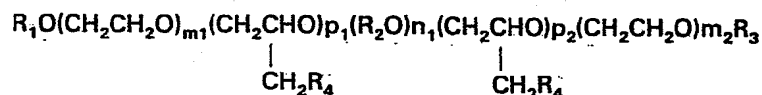
Namely, the present invention is characterized by incorporating into the photographic component layer of a silver halide photographic light-sensitive material for lithographic use a liquid or solid paraffin and a polyalkylene oxide-type compound, whereby a photographic-characteristically excellent silver halide photographic light-sensitive material having an extremely high contrast can be obtained which has been hardly solved by conventional techniques.

PREFERRED EMBODIMENT

In the present invention, preferred compounds usable in combination with liquid or solid paraffin

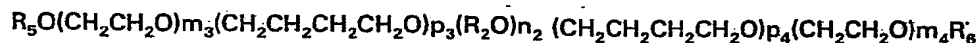
contained in aforesaid photographic component layer and having in the molecule thereof a polyalkylene oxide chain are those having preferably the Formulas (1), (2) and (3):

Formula (1)



5 Formula (2)

5

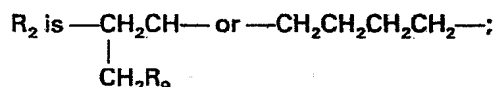


Formula (3)



10 wherein R_1 , R_3 , R_5 , R_6 , R_7 and R_8 each is hydrogen, a substituted or unsubstituted alkyl, aryl or acyl radical;

10

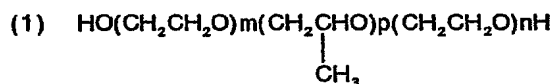


15 R_4 and R_9 each is hydrogen, an alkyl or cycloalkyl radical having from 1 to 4 carbon atoms; or an alkoxy or aryloxy radical having from 1 to 8 carbon atoms; n_1 , n_2 and n_3 each is an integer of up to 5; $P_1 + P_2$, $P_3 + P_4$ each is an integer of 0 to 60, preferably 0 to 30; and $m_1 + m_2$, $m_3 + m_4$ and m_5 each is an integer of from 10 to 80.

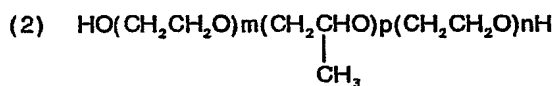
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The following are typical examples of those compounds having the above formulas, but the present invention is not limited thereto.

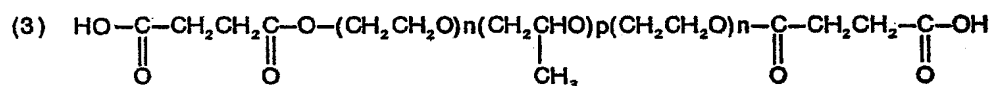
Exemplified compounds:



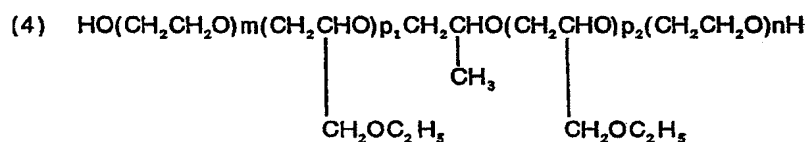
$$(m + n = 19 \quad p = 21)$$



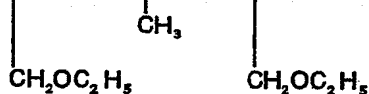
$$(m + n = 31 \quad p = 35)$$



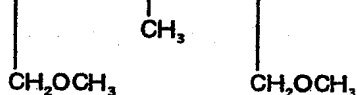
$$(m + n = 16 \quad p = 17)$$



$$(m + n = 24 \quad p_1 + p_2 = 12)$$



$$(m + n = 20 \quad p_1 + p_2 = 9)$$



$$(m + n = 22 \quad p_1 + p_2 = 20)$$



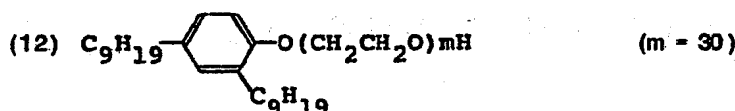
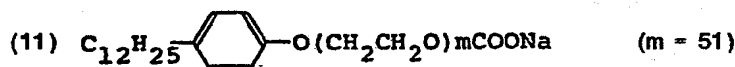
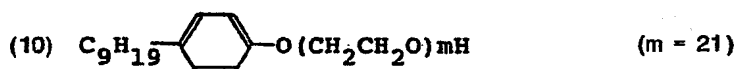
$$(m + n = 24 \quad p = 13)$$



$$(m + n = 18 \quad p = 7)$$



$$(m + n = 17 \quad p = 15)$$



According to the present invention, the amount of the abovementioned polyalkylene oxide-type compound to be added to the photographic component layer is within the range of from 0.01 g to 2.0 g per mole of silver halide, and more preferably, 0.03 g to 1.0 g. And the solvent to be used at the time of the adding is water or an organic solvent being mixable with water, for example, ethanol, methanol,

The foregoing solid paraffin usable in the photographic component layer in this invention, although desirable to be of the particle size of not more than $5\ \mu$, is particularly desired to be of the particle size of not more than $1\ \mu$. Namely, the particle size of the solid paraffin exceeding $5\ \mu$ can of course give an effect of the invention to some extent, but as the particle size becomes larger, not only does the stability of the paraffin-dispersed liquid or paraffin coating liquid become worse with the lapse of time but also the transparency of the resulting coat becomes deteriorated.

The liquid or solid paraffin of the present invention may be contained in the dispersed liquid form in the photographic component layer. The preparation of a dispersed liquid of the above-particle size-having solid paraffin is carried out by first dissolving the solid paraffin into a low-boiling solvent and then dispersing the resulting solution by means of, e.g., a homogenizer, into an aqueous solution of a protective colloid such as gelatin and a surface active agent. The above-mentioned low-boiling solvent, even if remaining in the dispersed liquid, will not reduce the effect of the present invention but is desirable to be removed either by heating or by evaporating under reduced pressure after completion of the dispersing operation.

As for the adding amount of the paraffin to be contained in the photographic component layer in this invention, although with the increase in the adding amount the effect becomes increasing, the fact is that an excessively large amount of the paraffin can cause the physical properties of the component layer to be deteriorated, so that the adding amount of the paraffin is desirable to be normally from 5% to 100% by weight, and particularly from 10% to 50% by weight to the hydrophilic binder content of the paraffin-containing layer.

No special restrictions are placed on the point of time for adding the foregoing liquid or solid paraffin and polyalkylene oxide-type compound used in this invention to the photographic component layer, but they are desirable to be added, for example, upon the formation of silver halide particles or upon completion of the second ripening process, and the photographic component layer to which they should be added is most preferably the emulsion layer containing silver halide, but the same effect can also be obtained even if they are added to other layers adjacent thereto or to an auxiliary layer or interlayer on the same side of the emulsion layer to the support.

According to the present invention, both the foregoing paraffin and the polyalkylene oxide-type compound may be incorporated together in a same layer but may also be incorporated separately in different layers.

For example, paraffin may be contained in the emulsion layer containing silver halide and polyalkylene oxide-type compound may be contained in an adjacent layer thereof, or, to the contrary, it is possible to cause the emulsion layer to contain polyalkylene oxide-type compound and cause the adjacent layer to contain paraffin respectively.

The effect of the present invention composed in the foregoing manner will be described in detail as follows.

Namely, those polyalkylene oxide-type compounds having the foregoing general formula, particularly polyoxyethylene-type compounds, display a development accelerative effect when present in a non-lith-type developer liquid, but on the contrary, display a development restraining effect when present in a lith-type developer liquid. This peculiar behavior is the fact well-known to those skilled in the art. This lith development restraining effect, when the light-sensitive material is processed in a lith-type developer liquid, gives satisfactory photographic characteristics, but on the contrary, when processed in a non-lith-type developer liquid, gives an undesirable development accelerative effect as described above, which therefore increases fog or reduces the contrast, thus making it difficult to obtain satisfactory photographic characteristics. Generally, in order to cope with this problem, an antifoggant is added, but most of antifoggants reduce not only fog but also the photographic speed or the contrast of the light-sensitive material as well as restrain the lith development itself, so that the amount of the alkylene oxide-type compound is compelled to be reduced, thus resulting in the deterioration of the photographic characteristics in the lith-type developer liquid.

The effect of the present invention can be displayed only by using the liquid or solid paraffin additionally to the previously described disadvantage-having polyalkylene oxide-type compound. And this combined use causes no recognizable increase in fog in the non-lith-type development and, besides, remarkably improves the contrast in a developer liquid, whether of the lith type or of the non-lith type, thus remarkably improving the photographic characteristics.

Following is a detailed description of the present invention. The composition of the silver halide used in the aforesaid emulsion layer containing silver halide in this invention, because the development speed needs to be rapid if an explosive development like the lith development is desired, is required to contain silver chloride in a quantity of not less than 50 mole% in the entire silver halide, and more preferably the silver halide used should be silver chlorobromide or silver chloriodobromide comprising not less than 60 mole% of silver chloride, not more than 40 mole% of silver bromide and not more than 5 mole% of silver iodide. These silver halides may be prepared by the neutral method, acid method, ammoniacal method, orderly mixing method, reverse mixing method, single-jet method, double-jet method, controlled double-jet method, conversion method, core-shell method, or the like, as described in "The Theory of the Photographic Process" by James, 4th ed., pp 88—104, published by McMillan Publishing Co. in 1977.

No particular restrictions are put on the particle size, granularity distribution, crystal habit, configuration (normal or twin, etc.) and the like, of the silver halide used, but it is desirable to use relatively uniformly distributed particle size-having silver halide with the size of not more than 1 μ . These silver halide particles or silver halide emulsions may also contain an iridium salt and/or rhodium salt for the purpose of improving the characteristics thereof to electronic flash light.

These silver halides, generally, may be chemically sensitized by the single or combined use of sulfur sensitizers such as sodium thiosulfate, thiourea, and the like; noble-metallic sensitizers including gold sensitizers such as chloroaurates, gold trichloride, etc., palladium sensitizers such as palladium chloride, chloropalladates, etc., platinum compounds, iridium compounds, and the like; selenium sensitizers such as selenious acid, selenourea, and the like; reduction sensitizers such as stannous chloride, polyamines such as diethylenetriamine, sulfites, silver nitrate, and the like; and may also be optically sensitized to any desired wavelength region by the single or combined use of cyanine dyes, merocyanine dyes, and the like.

In the present invention, there may be used such an electron-trapping compound as a metallic doping agent or desensitizing dye, which may also be applied to a direct-positive-type photographic silver halide emulsion containing not less than 50 mole% of fogged silver chloride.

As the vehicle for the above composition-having silver halide, gelatin, gelatin derivatives, synthetic hydrophilic polymers, or the like, may be used.

The silver halide emulsion layer for use in the present invention may contain various photographic additives: As antifoggants, for example, azaindenes such as 4-hydroxy-6-methyl-1,3,3a,7-tetraindene, triazoles, thiazoles, tetrazoles and other antifoggants known to those in the art may be used. As hardening agents, aldehyde compounds, ketone compounds, halogen-substituted acids such as mucochloric acid, ethyleneimine compounds, and the like, may be used. As coating aids, saponin, lauryl- or oleyl-monoethers or polyethylene glycol, and the like, may be used. As development accelerators, although not particularly restricted, such compounds as disclosed in, e.g., Japanese Patent O.P.I. Publication No. 24427/1974, quaternary ammonium salts, and the like, may be used. As physical property improving agents, those polymer latexes comprising homopolymers or copolymers of alkyl acrylates, alkyl methacrylates, acrylic acid, and the like, may be contained. And the silver halide emulsion layer or other light-sensitive material component layers of this invention may further contain such an antistatic agent as disclosed in, e.g., Japanese Patent Application No. 104940/1977, Japanese Patent O.P.I. Publication Nos. 56220/1976 and 46733/1974, and the like.

For the support to be used for coating thereon the photographic component layer in this invention, any arbitrary film such as, e.g., polyethylene terephthalate film, polycarbonate film, polystyrene film, cellulose acetate film, and the like, may be used.

It is preferable that the silver halide photographic light-sensitive material of the present invention is developed with the use of lith-type developing liquid though it may be developed with the use of developing liquid to be used for ordinary silver halide photographic light-sensitive material. Primary ingredients of the lith-type developing liquid consist of single or mixed components of developing agent in p-dihydroxybenzene type such as hydroquinone, alkyl-substituted hydroquinone and halogen-substituted hydroquinone etc. The amount of developing agent to be added is from 1 g to 100 g per 1 l of developing liquid and it preferably is from 10 g to 30 g.

The developing liquid may further contain following components and selected amounts of these components may be contained. Such component includes first the preservative of developing agent and as said preservative, sulfite (alkaline metal sulfite, alkaline metal metabisulfite) is used and its amount of not more than 6 g per 1 l of developing liquid is preferable to be used though there is no restriction in the concentration of free sulfite ion. Sulfite ion may further be added as a buffer and aldehyde-alkalhydrogen sulfite additives, additives of soluble sulfite and a compound having carbonyl ($>C=O$) such as additives of soluble sulfurous acid and cyclohexanone or acetone and further the mixture of aforesaid additives and aliphatic primary or aliphatic secondary amine group etc. are used as aforesaid buffer and the amount of sulfite ion buffer to be added is from 0 g to 150 g per 1 l of developing liquid and it preferably is from 20 g to 80 g. Alkaline agent may further be added to the lith-type developing liquid and hydroxide of alkaline metal, alkaline metal carbonate and alkaline metal borate etc. are used as aforesaid alkaline agent and a pH value of developing liquid is pH 8 or more, especially, it is preferable to be in the range from pH 9 to pH 11.

Aforesaid lith-type developing liquid may further contain at need the additives such as pH controlling agent, development controlling agent, organic antifoggant, antioxidant, development accelerator and organic solvent etc.

On the other hand, the developing agent for the developing liquid to be used for an ordinary silver halide photographic light-sensitive material includes hydroquinone, chlorohydroquinone, dihydroquinobenzene group such as catechol, 1-phenyl-3-pyrazolidone, 1-phenyl-4,4-dimethyl-3-pyrazolidone, 1-phenyl-4-methyl-4-hydroxymethyl-3-pyrazolidone, 3-pyrazolidone group such as 1-phenyl-4-methyl-4-hydroxymethyl-3-pyrazolidone, N-methyl-p-aminophenol, paraaminophenol group such as N-(4-hydroxyphenyl)glycine, β -methanesulfoneamidoethyl, ethylaminotoluidine, p-phenylenediamine group such as N,N-diethyl-p-phenylenediamine and ascorbic acid group, and an aqueous solution containing one or more of aforesaid developing agents is used and besides that, the

Composition b

| | | | |
|----|--|--------|----|
| | Triethyleneglycol | 9 g | |
| | 5-nitroindazole | 6 mg | |
| | Ethylenediaminedisodiumsalt tetraacetic acid | 6 g | |
| 5 | Boric acid | 2 g | 5 |
| | Potassium bromide | 2 g | |
| | Potassium carbonate | 50 g | |
| | Potassium sulfite | 2.5 g | |
| | Sodium hydroxide | 1 g | |
| 10 | Add water to make | 167 ml | 10 |

The mixing ratio of the above compositions to water for the use thereof is as follows.
Composition a: composition b: water = 1:1:4 (ratio by volume)

(Developing liquid-B)

| | | | |
|----|--|---------|----|
| | Ethylenediaminedisodiumsalt tetraacetic acid | 8 g | |
| 15 | 5-Nitroindazole | 1 g | 15 |
| | 1-phenyl-5-mercaptotetrazole | 0.36 g | |
| | Add potassium hydroxide to make | pH 11.3 | |
| | Potassium sulfite | 253 g | |
| | Potassium bromide | 13 g | |
| 20 | Hydroquinone | 80 g | 20 |
| | Potassium carbonate | 40 g | |
| | Diethyleneglycol | 100 g | |
| | 1-phenyl-4,4-dimethyl-3-pyrazolidinone | 1.4 g | |
| | Add pure water to make | 1 l | |

25

When said liquid B is used, pure water was added to make 4 l.

The results obtained by these processings are as shown in Table 1 wherein the contrast values represent the inclination of the toe of the characteristic curves considered particularly important.

25

TABLE 1

| Developer | A | | B | |
|------------------------------|------|----------|------|----------|
| Photographic characteristics | Fog | Contrast | Fog | Contrast |
| Sample-1 | 0.07 | 4.2 | 0.14 | 3.8 |
| Sample-2 | 0.05 | 5.8 | 0.19 | 3.2 |
| Sample-3 | 0.04 | 6.4 | 0.26 | 2.7 |
| Sample-4 | 0.04 | 6.8 | 0.11 | 3.6 |

As apparent from Table 1, the addition of an increased amount of the polyalkylene oxide-type compound to the light-sensitive material free of the paraffin of this invention is capable of improving the photographic characteristics in the lith-type development, but on the contrary deteriorates the photographic characteristics in the non-lith-type development. In contrast, the combined use of both the paraffin and the polyalkylene oxide-type compound improves the photographic characteristics in both the lith-type development and the non-lith-type development, i.e., an excellent contrast can be obtained without increasing fog.

EXAMPLE 2

To part of the remaining foregoing liquid A were added 700 mg per mole of silver halide of Exemplified Compound (9) to thereby obtain a mixture. The mixture was then divided into three parts. One of them to which was added no paraffin was regarded as sample-5, another to which was added the same amount of the liquid paraffin as in the foregoing sample-4 was regarded as sample-6, and still another to which was added the solid paraffin in the same manner was regarded as sample-7. These were coated and dried in the same manner as in Example 1, and then subjected to both lith-type and non-lith-type developments. The obtained results are as shown in Table 2.

There was observed from Table 2 above that paraffin of the invention displays excellent photographic characteristic improvements, even if it is liquid or solid paraffin.

EXAMPLE 3

To the finally remaining part of the foregoing Liquid A were added 400 mg per mole of silver halide of Exemplified Compound (4), and the resulting mixture was divided into five parts to prepare sample-8 containing no paraffin, sample-9 containing the liquid paraffin in the same amount as in the foregoing sample-4, sample-10 containing the liquid paraffin in the amount twice as much as that used in sample-9, sample-11 containing 50 mg per mole of silver halide of 5-nitrobenzimidazole as an antifoggant, and sample-12 containing the 5-nitrobenzimidazole in the amount twice as much as that used in sample-11. These were coated, dried and processed in the same manner as in Example 1. The obtained results are as shown in Table 3.

TABLE 2

| Developer | A | | B | |
|------------------------------|------|----------|------|----------|
| Photographic characteristics | Fog | Contrast | Fog | Contrast |
| Sample-5 | 0.06 | 6.4 | 0.20 | 2.9 |
| Sample-6 | 0.04 | 6.7 | 0.07 | 3.9 |
| Sample-7 | 0.04 | 6.6 | 0.09 | 3.7 |

TABLE 3

| Developer | A | | | B | | |
|------------------------------|------|----------|----------------|------|----------|----------------|
| Photographic characteristics | Fog | Contrast | Relative speed | Fog | Contrast | Relative speed |
| Sample-8 | 0.05 | 6.5 | 100 | 0.19 | 2.7 | 176 |
| Sample-9 | 0.04 | 6.7 | 103 | 0.09 | 3.5 | 179 |
| Sample-10 | 0.04 | 6.8 | 101 | 0.06 | 3.7 | 180 |
| Sample-11 | 0.04 | 5.7 | 83 | 0.11 | 2.4 | 155 |
| Sample-12 | 0.04 | 4.9 | 61 | 0.06 | 2.1 | 128 |

According to Table 3, the additional use of 5-nitrobenzimidazole as an antifoggant to the samples of the combined use of this invention displays a fog-restraining effect but has a reverse effect upon the photographic speed or the contrast. In contrast to this, the combined use of the paraffin and the polyalkylene oxide-type compound of this invention well restrains fog with little or no influence upon the photographic speed and, in addition, shows an excellent contrast effect.

CLAIMS

1. A silver halide photographic light-sensitive material comprising on a support:

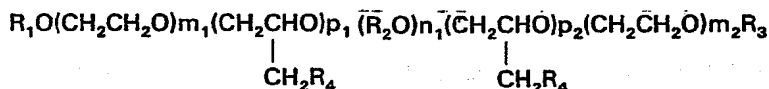
(1) silver halide grains containing at least 50 mole% of silver chloride;

(2) a liquid paraffin or finely particulate solid paraffin having an average particle size of not more than $5\ \mu$;

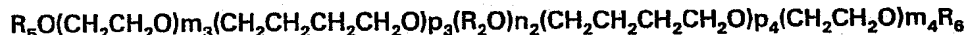
(3) a compound comprising an alkylene oxide chain present in one or more layers.

2. A material according to claim 1, wherein said compound has the formula (1), (2) or (3):

Formula (1)



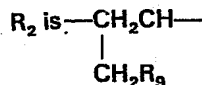
Formula (2)



Formula (3)



(wherein R_1 , R_3 , R_5 , R_6 , R_7 and R_8 represent hydrogen or an alkyl, aryl or acyl radical;



or $\text{---CH}_2CH_2CH_2CH_2\text{---}$;

R_4 and R_9 independently represent hydrogen, an alkyl or cycloalkyl radical having from 1 to 4 carbon atoms, or an alkoxy radical having from 1 to 8 carbon atoms; n_1 , n_2 and n_3 independently represent 0 or an integer from 1 to 5; $p_1 + p_2$, $p_3 + p_4$ represent 0 or an integer from 1 to 60; and $m_1 + m_2$, $m_3 + m_4$ and m_5 represent an integer of from 10 to 80.

3. A material according to claim 2, wherein $p_1 + p_2$, $p_3 + p_4$ represent 0 or an integer from 1 to 30.

4. A material according to claim 1, 2 or 3 wherein said compound is present in the amount from 0.01 g to 2.0 g per mole of silver halide.

5. A material according to any one of claims 1 to 4, wherein said paraffin is present in an amount from 5% to 100% by weight based on the weight of hydrophilic binder.

6. A material according to any one of claims 1 to 5, wherein said silver halide grains have an average size of 0.1 to 1 μ .
7. A material according to any one of claims 1 to 6, wherein said paraffin and said compound are contained in different layers.
- 5 8. A material according to any one of claims 1 to 6, wherein said paraffin and said compound are contained in the same layer. 5
9. A material according to any one of claims 1 to 8, wherein said silver halide grains consist of silver chloride, silver bromide and silver iodide.
- 10 10. A material according to any one of claims 1 to 8, wherein said silver halide grains consist of silver chloride and silver bromide. 10
11. A material according to any one of claims 1 to 10, wherein said solid particulate paraffin has an average size of 0.05 to 5 μ .
12. A material according to any one of claims 1 to 11, wherein the said compound is one specifically identified herein.
- 15 13. A material according to claim 1 substantially as described in any one of the Examples. 15

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